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(54) **Acaricidal composition.**

(57) An acaricidal composition comprises, as active ingredient, one or more compounds selected from methyl cinnamate, ethyl cinnamate, n-propyl cinnamate, isopropyl cinnamate, n-butyl cinnamate, isobutyl cinnamate, isoamyl cinnamate, n-hexyl cinnamate, allyl cinnamate, cinnamyl acetate, cinnamyl propionate, cinnamyl n-butyrate, cinnamyl isobutyrate, p-cresyl acetate, p-cresyl butyrate, p-cresyl isobutyrate, p-methylbenzyl propionate, β-phenoxyethyl alcohol, phenoxyethyl acetate phenoxyethyl propionate, phenoxyethyl n-butyrate, phenoxyethyl isobutyrate, methyl phenylacetate, ethyl phenylacetate, dibenzyl ether, heliotropin, methyl diphenyl ether and 2-methyl-1-(methylbicyclo[2.2.1]hept-5-en-2-yl)-1-penten-3-ol.

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ACARICIDAL COMPOSITION

This invention relates to an acaricidal composition which is free from harmful effects on human beings and is very effective for exterminating house dust acari.

House dust acari inhabit, and propagate mainly in, highly moist places, for example, on the surface of floors, under or within floor coverings such as tatami or carpet, or within bedclothes. Recently, Dermatophagoides including Dermatophagoides pteronyssinus and Dermatophagoides farinae, which constitute 90% of house dust acari, have become a serious problem since they are important allergens causing bronchial asthma, allergic rhinitis and atopic dermatitis.

The most effective method for exterminating these acari is to ventilate and dry the house well. However, the recent increase in the number of houses having a closed structure and changes in life style make it more and more difficult to ventilate a room sufficiently. Under these circumstances, the damage caused by acari has become more and more serious.

In order to exterminate these acari, various acaricides (for example organophosphorus compounds such as fenitrothion, fenthion, dichlorvos, diazinon; carbamate compounds such as propoxur, carbaryl; pyrethroid compounds such as resmethrin, phenothrin, permethrin) have been applied in the form of aerosol, fumigant, insecticidal sheet or impregnating agent for, eg, carpets. Furthermore, it was recently proposed to use compounds other than those cited above for exterminating acari. For example, JP-A-61-57501 discloses using a combination of acaricidal compounds such as benzyl benzoate, benzyl salicylate or dibutyl phthalate with a powdery cleanser, and indicates that the acaricidal effect of benzyl benzoate has been physiologically particularly well studied. (The term "JP-A" as used herein means an "unexamined published patent application".) JP-A-61-91103 discloses an acaricide which comprises benzyl benzoate and an aliphatic hydrocarbon as the major components. Further, JP-A-61-87603 discloses benzyl salicylate and phenethyl benzoate, while JP-A-62-33106 discloses phenyl salicylate, phenyl benzoate, diphenylamine, methyl β-naphthyl ketone and coumarin each as an active ingredient for an acaricide. Furthermore, JP-A-64-19004 discloses an acaricide comprising benzaldehyde or perillaldehyde, l-carvone or d-carvone, methyl salicylate or ethyl salicylate, or methyl benzoate or ethyl benzoate as an active ingredient. Regarding natural substances, furthermore, JP-A-63-104905 discloses that terpene compounds are available as acari-prevention agents. Furthermore, it is known that other vegetable essential oils (for example, bitter almond oil, wintergreen oil) show an acaricidal effect (F. Watanabe et al., Shoyakugaku Zasshi, 43 [2], 163-168 (1989)).

However, typical known acaricidal compounds (particularly organophosphorus compounds and carbamate compounds) generally show a high toxicity and exert undesirable effects on the human body. Therefore, it is undesirable to use these compounds in confined conditions or around houses. These compounds are further disadvantageous in that their effects on Dermatophagoides causing allergic diseases are limited. On the other hand, pyrethroid compounds are expensive and show only limited effects on house dust acari, though they are less toxic in general. Other acaricidal compounds are also disadvantageous in their limited effects on Dermatophagoides.

Accordingly, it has been urgently required to develop an acaricide which is very safe with respect to effects on the human body, can be easily used anywhere in the house, and yet exerts a powerful effect in exterminating a number of house dust acari, including Dermatophagoides, which cause allergic diseases.

We have found that the following compounds, which have been used as perfumes in foods and cosmetics for a long time and have thus been proved to be harmless to human beings, are highly effective in the extermination of house dust acari.

According to the present invention there is provided an acaricidal composition comprising as the active ingredient one or more compounds selected from among methyl cinnamate, ethyl cinnamate, n-propyl cinnamate, isopropyl cinnamate, n-butyl cinnamate, isobutyl cinnamate, isoamyl cinnamate, n-hexyl cinnamate, allyl cinnamate, cinnamyl acetate, cinnamyl propionate, cinnamyl n-butyrate, cinnamyl isobutyrate, p-cresyl acetate, p-cresyl butyrate, p-cresyl isobutyrate, p-methylbenzyl propionate, β-phenoxyethyl alcohol, phenoxyethyl acetate, phenoxyethyl propionate, phenoxyethyl n-butyrate, phenoxyethyl isobutyrate, methyl phenylacetate, ethyl phenylacetate, dibenzyl ether, heliotropin, methyl diphenyl ether and 2-methyl-1-(methylbicyclo[2.2.1]hept-5-en-2-yl)-1-penten-3-ol.

The acari to be exterminated with the acaricidal composition of the present invention include not only house dust acari inhabiting and propagating indoors, for example, Pyroglyphidae such as Dermatophagoides farinae and Dermatophagoides pteronyssinus; Acaridae such as Tyrophagus putrescentiae and Aleuroglyphus ovatus; Glycyphagidae such as Glycyphagus privatus and Glycyphagus domesticus; and Cheyletidae such as Cheyletus malaccensis and Cheyletus fortis; but animal-parasitic acari, for example,

Marcronyssidae such as Ornithonyssus bacoti and Ormithonyssus sylviarum.

The acaricidal composition of the present invention may consist of one of the above-mentioned active ingredients or a combination thereof, as such. In general, however, it may be formulated into an oil preparation, emulsifiable concentrate, wettable powder, spray, aerosol, fumigant, coating, detergent, dust, granules or capsules by supporting on a solid or liquid carrier and optionally adding various additives, for example, film-forming agent, emulsifier, sticking agent, dispersant, wetting agent, stabilizer, propellant and volatility-controller, if required.

Examples of the solid carrier to be used herein include mineral powders such as silicic acid, kaolin, activated carbon, bentonite, diatomaceous earth, talc and calcium carbonate; vegetable powders such as wheat flour and starch; and synthetic polymer powder such as polyvinyl chloride powder. Examples of the liquid carrier include water; aliphatic hydrocarbons such as hexane, kerosene and coal oil; aromatic hydrocarbons such as benzene, toluene and xylene; halogenated hydrocarbons such as dichloroethane and carbon tetrachloride; alcohols such as ethanol, isopropyl alcohol and ethylene glycol; ketones such as acetone, methyl ethyl ketone and cyclohexanone; others such as tetrahydrofuran, dimethoxyethane and diethyl ether; esters such as ethyl acetate; nitriles such as acetonitrile; acid amides such as dimethylformamide; and vegetable oils such as soybean oil and cotton seed oil.

Examples of the film-forming agent include cellulose derivatives, vinyl resins, alkyd resins, urea resins, epoxy resins, polyester resins, urethane resins, silicone resins, acrylic resins, chlorinated rubbers and polyvinyl alcohol. Examples of the emulsifier, sticking agent and dispersant include surfactants such as alkyl polyoxyethylene alkylaryl ethers, polyoxyethylene fatty acid esters, fatty acid glycerols, sorbitan fatty acid esters, higher alcohol sulfates and alkylarylsulfonic acid salts. Examples of the propellant include liquified petroleum gas, Freon gas and dimethyl ether. Examples of the volatility-controller include tricyclodecane and cyclododecane.

Furthermore, the active ingredient(s) may be used together with sublimating insecticides such as paradichlorobenzene, naphthalene or camphor so as to give a sublimating solid preparation.

Moreover, the acaricidal composition of the present invention may contain, for example, various conventional insecticides, acaricides, synergists, harmful insect repellents, rodent repellents, bactericides, fungicides, perfumes or colorants used for exterminating harmful insects such as fenitrothion, propoxur or resmethrin.

The content of the above-mentioned active ingredient in the acaricidal composition of the present invention may vary depending on the formulation, application means and the place to be applied. It is generally preferable that the total content of the active ingredient(s) ranges from 0.1 to 50% by weight (in the case of wettable powder or emulsifiable concentrate) and from 0.1 to 30% by weight (in the case of oil preparation or aerosol), respectively.

The acaricidal composition of the present invention thus prepared may be applied to, for example, floor, *tatami*, carpets, bedclothes, sofas, pillows or closets by depositing, spraying, coating, transpiring or placement. Alternatively, it may be used as a detergent for human or pet animals. The dose is preferably approximately 80 mg or more per m^2 of the area to be treated or approximately 8 mg or more per m^3 of the space to be treated, in terms of the total amount of the active ingredient.

In addition to the above formulations, the acaricide of the present invention may be formulated into film, sheet or constructional material having an acaricidal activity by supporting the active ingredient(s) on an appropriate substrate. Examples of the substrate to be used herein include sheets of synthetic resins such as polyethylene, polypropylene, nylon, polyvinyl chloride or polyesters; animal or vegetable fibrous materials or inorganic fibrous materials such as paper, cloth, non-woven cloth and leather; mixed sheets of the above-mentioned synthetic resins and animal, vegetable or inorganic fibers; mixed fabrics or non-woven fabrics; foils or films of metals such as aluminum, stainless steel or zinc; laminates of the above-mentioned sheets; and various natural wooden materials and plastics molded articles employed for constructional purposes. The active ingredient of the acaricidal composition of the present invention is supported on these substrates by coating, impregnating, depositing or fabricating to give an acaricidal material. The amount of the active ingredient in the substrate is not particularly restricted but may be optionally selected. In the case of impregnation, it is preferable to use the active ingredient in the saturation amount.

The acaricidal material thus obtained may be preferably used, for example, in the following manner. A polymer sheet (for example, polypropylene) impregnated with the active ingredient of the present invention is placed under *tatami*, carpets or sofas. In this case, it is preferable to use the active ingredient at a ratio of from approximately 0.5 to 20 g per unit area. The impregnation of the polymer with the active ingredient makes the sustained release of the active ingredient possible, which brings about a sustained acaricidal effect.

The effects of the active ingredients of the present invention were examined by using Dermatophagoides pteronyssinus, which is one of Dermatophagoides and is generally less sensitive to chemicals, by the following procedure.

Namely, a filter paper (5 mm x 5 mm) is impregnated with each test compound in such a manner as to give the definite concentration. A liquid compound is used as such while a solid one is dissolved in acetone. In accordance with a method reported by Watanabe et al., Shoyakugaku Zasshi, 43 [2], 163-168 (1989) the filter sheet is introduced into a cylindrical container (approximately 20 cc) containing 50 to 80 head of Dermatophagoides pteronyssinus together with a bait. The container is then sealed with a Teflon stopper and allowed to stand in an incubator at 25° C. After 24 hours and 48 hours, the life or death of the acari is examined under a stereoscopic microscope or a loupe (x 25) and evaluated. The procedure is repeated thrice and the lethality is calculated according to the following equation. Table 1 shows average values.

Lethality (%) = (X - Y)/X x 100

X: number of living acari in untreated plot; and

Y: number of living acari in treated plot.

In Table 1, a mixture of Test Compounds is expresed by the Compound Number of each compcnent.

For comparison, permethrin and benzyl salicylate, which are conventional acaricides, were also evaluated in the same manner. The results are shown in Table 1.

TABLE 1

Type	Compound No.	Test Compound (blending ratio)	Lethality: Dose of active ingredient (0.08 g/m²) After 24 hours	Lethality: Dose of active ingredient (0.08 g/m²) After 48 hours	Lethality: Dose of active ingredient (0.04 g/m²) After 24 hours	Lethality: Dose of active ingredient (0.04 g/m²) After 48 hours
Single compound	(1)	Methyl cinnamate	100	100	100	100
	(2)	Ethyl cinnamate	100	100	100	100
	(3)	n-Propyl cinnamate	86	100	78	90
	(4)	Isopropyl cinnamate	92	100	82	98
	(5)	n-Butyl cinnamate	80	100	75	90
	(6)	Isobutyl cinnamate	82	100	71	75
	(7)	Isoamyl cinnamate	88	100	76	100
	(8)	n-Hexyl cinnamate	82	100	58	62
	(9)	Allyl cinnamate	79	100	72	100
	(10)	Cinnamyl acetate	100	100	53	82
	(11)	Cinnamyl propionate	100	100	65	89
	(12)	Cinnamyl n-butyrate	80	100	53	72
	(13)	Cinnamyl isobutyrate	100	100	62	85
	(14)	p-Cresyl acetate	85	100	80	98

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TABLE 1 (cont'd)

Type	Compound No.	Test Compound (blending ratio)	Lethality: Dose of active ingredient (0.08 g/m²) After 24 hours	Lethality: Dose of active ingredient (0.08 g/m²) After 48 hours	Lethality: Dose of active ingredient (0.04 g/m²) After 24 hours	Lethality: Dose of active ingredient (0.04 g/m²) After 48 hours
Single compound	(15)	p-Cresyl butyrate	92	100	83	97
	(16)	p-Cresyl isobutyrate	90	100	78	85
	(17)	p-Methylbenzyl propionate	95	100	86	96
	(18)	β-Phenoxyethyl alcohol	100	100	96	100
	(19)	Phenoxyethyl acetate	100	100	86	98
	(20)	Phenoxyethyl propionate	100	100	78	85
	(21)	Phenoxyethyl n-butyrate	100	100	82	98
	(22)	Phenoxyethyl isobutyrate	100	100	100	100
	(23)	Methyl phenylacetate	100	100	100	100
	(24)	Ethyl phenylacetate	100	100	100	100
	(25)	Dibenzyl ether	100	100	100	100
	(26)	Heliotropin	100	100	100	100
	(27)	Methyl diphenyl ether	100	100	100	100

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TABLE 1 (cont'd)

Type	Compound No.	Test Compound (blending ratio)	Lethality: Dose of active ingredient (0.08 g/m²) After 24 hours	Lethality: Dose of active ingredient (0.08 g/m²) After 48 hours	Lethality: Dose of active ingredient (0.04 g/m²) After 24 hours	Lethality: Dose of active ingredient (0.04 g/m²) After 48 hours
Single compound	(28)	2 Methyl-1-(methylbicyclo[2.2.1]hept-5-en-2-yl)-1-penten-3-ol	100	100	69	100
Mixed Composition		(2)/(15) (1/1)	100	100	100	100
		(2)/(18) (1/1)	100	100	100	100
		(2)/(24) (1/1)	100	100	100	100
		(2)/(27) (1/1)	100	100	100	100
		(14)/(18) (1/1)	100	100	98	100
		(14)/(23) (1/1)	100	100	90	100
		(14)/(25) (1/1)	100	100	100	100
		(14)/(27) (1/1)	100	100	100	100
		(2)/(18)/(24) (1/1/1)	100	100	92	100
		(24)/(25)/(27) (1/1/1)	100	100	100	100
		(25)/(26)/(27) (1/1/1)	100	100	90	100

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TABLE 1 (cont'd)

Type	Compound No.	Test Compound (blending ratio)	Lathality: Dose of active ingredient (0.08 g/m²) After 24 hours	Lathality: Dose of active ingredient (0.08 g/m²) After 48 hours	Lathality: Dose of active ingredient (0.04 g/m²) After 24 hours	Lathality: Dose of active ingredient (0.04 g/m²) After 48 hours
Comparison		Permethrin	82	90	63	83
		Benzyl salicylate	50	70	25	32

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As Table 1 clearly shows, the active ingredients of the acaricide of the present invention were superior to permethrin and benzyl salicylate for exterminating Dermatophagoides pteronyssinus.

To further illustrate the present invention, and not by way of limitation, the following Examples will be given.

EXAMPLE 1

Oil Preparation:	
	(parts by weight)
Ethyl cinnamate	2
Isopropyl alcohol	98
Total	100

The above components were mixed under stirring to give a homogeneous oil preparation.

EXAMPLE 2

Emulsifiable Concentrate:	
	(parts by weight)
Cinnamyl acetate	20
Sorbitan monostearate	10
Xylene	70
Total	100

The above components were mixed under stirring to give a homogeneous emulsion.

EXAMPLE 3

Dust:	
	(parts by weight)
β-Phenoxyethyl alcohol	10
Silicic anhydride	5
Talc	85
Total	100

The above components were intimately mixed to give a homogeneous dust.

EXAMPLE 4

Dust:	
	(parts by weight)
Methyl phenylacetate	40
Soft polyvinyl chloride powder	60
Total	100

The above components were stirred at room temperature over day and night to allow the polyvinyl chloride powder to absorb the methyl phenylacetate. Thus a dust was prepared.

EXAMPLE 5

Detergent:	
	(parts by weight)
p-Cresyl butyrate	10
Polyoxyethylene monylphenyl ether	25
water	65
Total	100

The above components were intimately mixed to give a homogeneous detergent.

EXAMPLE 6

Aerosol:	
	(parts by weight)
Ethyl phenylacetate	10
Dimethoxyethane	40
Liquefied petroleum gas	50
Total	100

The ethyl phenylacetate and dimethoxyethane were mixed under stirring and then introduced into an aerosol container. After providing a valve, the liquefied petroleum gas was fed thereinto through the valve under a pressure to give an aerosol.

EXAMPLE 7

Aerosol:	
	(parts by weight)
p-Cresyl butyrate	5
Methyl diphenyl ether	5
Xylene	10
Illuminating kerosene	30
Liquefied petroleum gas/dimethyl ether mixture (ratio by volume = 1:1)	50
Total	100

The above components except the mixture of liquefied petroleum gas and dimethyl ether were mixed under stirring and then introduced into an aerosol container. After providing a valve, the mixture of liquefied petroleum gas and dimethyl ether was fed thereinto through the valve under a pressure to give an aerosol.

EXAMPLE 8

Sheet material:	
	(parts by weight)
Methyl phenylacetate	20
Ethyl cellulose	10
Ethanol	70
Total	100

The above components were mixed under stirring, and a polyethylene pulp non-woven fabric was impregnated therewith in such a manner as to give a ratio of methyl pheny acetate of 1 g/m^2. Thus a sheet material was obtained.

EXAMPLE 9

Sheet material:	
	(parts by weight)
Ethyl cinnamate	10
Dibenzyl ether	10
Ethyl cellulose	10
Ethanol	70
Total	100

The above components were mixed under stirring, and a polyethylene pulp non-woven fabric was impregnated therewith in such a manner as to give a total amount of ethyl cinnamate and dibenzyl ether of 1 g/m^2. Thus a sheet material was obtained.

The acaricidal composition of the present invention exhibits an excellent effect of extermination house dust acari. Further, it is highly safe to human body and can be easily applied in the house, which makes it extremely advantageous.

Claims

1. An acaricidal composition comprising one or more compounds selected from methyl cinnamate, ethyl cinnamate, n-propyl cinnamate, isopropyl cinnamate, n-butyl cinnamate, isobutyl cinnamate, isoamyl cinnamate, n-hexyl cinnamate, allyl cinnamate, cinnamyl acetate, cinnamyl propionate, cinnamyl n-butyrate, cinnamyl isobutyrate, p-cresyl acetate, p-cresyl butyrate, p-cresyl isobutyrate, p-methylbenzyl propionate, β-phenoxyethyl alcohol, phenoxyethyl acetate, phenoxyethyl propionate, phenoxyethyl n-butyrate, phenoxyethyl isobutyrate, methyl phenyl-acetate, ethyl phenylacetate, dibenzyl ether, heliotropin, methyl diphenyl ether and 2-methyl-1-(methylbicyclo[2.2.1]hept-5-en-2-yl)-1-penten-3-ol as an active ingredient.

2. An acaricidal composition as claimed in claim 1 and containing a solid or liquid carrier.

3. An acaricidal composition as claimed in claim 2, wherein the active ingredient is present in an amount of from 0.1 to 50% by weight.

4. An acaricidal composition as claimed in claim 3, wherein said composition is in the form of a wettable powder or an emulsifiable concentrate.

5. An acaricidal composition as claimed in claim 2, wherein the active ingredient is present in an amount of from 0.1 to 30% by weight.

6. An acaricidal composition as claimed in claim 5, wherein said composition is in the form of an oil preparation or an aerosol.

7. A method of exterminating house dust acari, which comprises applying an acaricidal composition comprising, as active ingredient, one or more compounds selected from methyl cinnamate, ethyl cinnamate, n-propyl cinnamate, isopropyl cinnamate, n-butyl cinnamate, isobutyl cinnamate, isoamyl cinnamate, n-hexyl cinnamate, allyl cinnamate, cinnamyl acetate, cinnamyl propionate, cinnamyl n-butyrate, cinnamyl isobutyrate, p-cresyl acetate, p-cresyl butyrate, p-cresyl isobutyrate, p-methylbenzyl propionate, β-phenoxyethyl alcohol, phenoxyethyl acetate, phenoxyethyl propionate, phenoxyethyl n-butyrate, phenoxyethyl isobutyrate, methyl phenylacetate, ethyl phenylacetate, dibenzyl ether, heliotropin, methyl diphenyl ether and 2-methyl-1-(methylbicyclo[2.2.1]hept-5-en-2-yl)-1-penten-3-ol to a location inhabited by house dust acari.

8. A method as claimed in claim 7, wherein said house dust acari are Dermatophagoides.

9. Use as an acaricide of any of the compounds listed in claim 1, either singly or in any combination thereof.

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EUROPEAN SEARCH REPORT

Application Number

EP 90 31 1484

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X	CHEMICAL ABSTRACTS, vol. 88, no. 3, 16th January 1978, page 163, abstract no. 17260z, Columbus, Ohio, US; R.S. DESHPANDE et al.: "Insecticidal activity of *Ocimum basilicum Linn*", & PESTICIDES 1977, 11(5), 11-12 * Abstract * ---	1	A 01 N 37/10 A 01 N 37/02 A 01 N 39/00 A 01 N 31/04 A 01 N 43/30 A 01 N 31/14
X	US-A-3 259 648 (H.E. HENNIS) * Whole document * ---	1-9	
A	EP-A-0 235 722 (BASF AG) ---		
X	FR-A-2 392 602 (BLOCK DRUG CO., INC.) * Page 1, lines 1-3; page 2, lines 15-19; tables, claims * ---	1-9	
X	US-A-4 368 207 (BLOCK DRUG CO., INC.) * Column 1, line 58 - column 2, line 18; tables; claims 1-11 * ---	1-9	
P,X	WO-A-8 912 673 (VAX APPLIANCES LTD) * Page 5, lines 6-13; page 21, lines 1-10; page 22, lines 3-11; claims 1-6 * ---	1-9	TECHNICAL FIELDS SEARCHED (Int. Cl.5) A 01 N
X	CHEMICAL ABSTRACTS, vol. 90, no. 19, 7th May 1979, page 169, abstract no. 147035g, Columbus, Ohio, US; & HU-A-15 661 (KOZMETIKAI ES HAZTARTASVEGYIPARI VALLALAT) 28-11-1978 * Abstract * --- -/-	1-9	

~~The present search report has been drawn up for all claims~~

Place of search	Date of completion of the search	Examiner
THE HAGUE	01-02-1991	DONOVAN T.M.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P0401)

European Patent Office

CLAIMS INCURRING FEES

The present European patent application comprised at the time of filing more than ten claims.

☐ All claims fees have been paid within the prescribed time limit. The present European search report has been drawn up for all claims.

☐ Only part of the claims fees have been paid within the prescribed time limit. The present European search report has been drawn up for the first ten claims and for those claims for which claims fees have been paid,

namely claims:

☐ No claims fees have been paid within the prescribed time limit. The present European search report has been drawn up for the first ten claims.

x LACK OF UNITY OF INVENTION

The Search Division considers that the present European patent application does not comply with the requirement of unity of invention and relates to several inventions or groups of inventions.

namely:

See sheet -B-

☐ All further search fees have been paid within the fixed time limit. The present European search report has been drawn up for all claims.

☒ Only part of the further search fees have been paid within the fixed time limit. The present European search report has been drawn up for those parts of the European patent application which relate to the inventions in respect of which search fees have been paid,

namely claims: points 1.,3.,4.,5., and 6.

☐ None of the further search fees has been paid within the fixed time limit. The present European search report has been drawn up for those parts of the European patent application which relate to the invention first mentioned in the claims,

namely claims:

European Patent Office

EUROPEAN SEARCH REPORT

Application Number

EP 90 31 1484

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.5)
X	DERWENT CENTRAL PATENTS INDEX, BASIC ABSTRACTS JOURNAL, section C: AGDOC, week E11, 12th May 1982, class C03, abstract no. 21087E/11, Derwent Publications Ltd, London, GB; & JP-A-57 024 303 (MITSUI TOATSU CHEM. INC.) 08-02-1982	1-9	
X	FR-A- 674 743 (IG FARBENINDUSTRIE AG) * Whole document *	1-9	
X	"The Merck Index", edition 10, 1983, Merck & Co., Inc., Rahway, NJ, US * Page 1078, compound no. 7350 *	1-9	
P,X	WO-A-9 009 738 (CHARWELL CONSUMER PRODUCTS LTD) * Whole document *	1-9	
			TECHNICAL FIELDS SEARCHED (Int. Cl.5)

~~The present search report has been drawn up for all claims~~

Place of search	Date of completion of the search	Examiner
THE HAGUE	01-02-1991	DONOVAN T.M.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
P : intermediate document

T : theory or principle underlying the invention
E : earlier patent document, but published on, or after the filing date
D : document cited in the application
L : document cited for other reasons

& : member of the same patent family, corresponding document

EPO FORM 1503 03.82 (P0401)

LACK OF UNITY OF INVENTION

The Search Division considers that the present European patent application does not comply with the requirement of unity of invention and relates to several inventions or groups of inventions, namely:

1. Claims 1-9 (partially), as far as the active ingredient is one or more of methyl cinnamate, ethyl cinnamate, n-propyl cinnamate, isopropyl cinnamate, n-butyl cinnamate, isobutyl cinnamate, isoamyl cinnamate, n-hexyl cinnamate, allyl cinnamate, cinnamyl acetate, cinnamyl propionate, cinnamyl n-butyrate or cinnamyl isobutyrate, optionally with one or more of p-cresyl acetate, p-cresyl butyrate, p-cresyl isobutyrate, p-methylbenzyl propionate, β-phenoxyethyl alcohol, phenoxyethyl acetate, pheroxyethyl propionate, phenoxyethyl n-butyrate, phenoxyethyl isobutyrate, methyl phenylacetate, ethyl phenylacetate, dibenzyl ether, heliotropin, methyl diphenyl ether or 2-methyl-1- methylbicyclo(2.2.1)hept-5-en-2-yl)-1-penten-3-ol.

2. Claims 1-9 (partially), as far as the active ingredient is one or more of p-cresyl acetate, p-cresyl butyrate, p-cresyl isobutyrate or p-methylbenzyl propionate, optionally with one or more of β-phenoxyethyl alcohol, phenoxyethyl acetate, phenoxyethyl propionate, phenoxyethyl n-butyrate, phenoxyethyl isobutyrate, methyl phenylacetate, ethyl phenylacetate, dibenzyl ether, heliotropin, methyl diphenyl ether or 2-methyl-1-(methylbicyclo(2.2.1)hept-5-en-2-yl)-1-penten-3-ol.

3. Claims 1-9 (partially), as far as the active ingredient is one or more of β-phenoxyethyl alcohol, phenoxyethyl acetate, phenoxyethyl propionate, phenoxyethyl n-butyrate or phenoxyethyl isobutyrate, optionally with one or more of methyl phenylacetate, ethyl phenylacetate, dibenzyl ether, heliotropin, methyl diphenyl ether or 2-methyl-1-(methyl-bicyclo(2.2.1)hept-5-en-2-yl)-1-penten-3-ol.

4. Claims 1-9 (partially), as far as the active ingredient is one or more of methyl phenylacetate or ethyl phenylacetate, optionally with one or more of dibenzyl ether, heliotropin, methyl diphenyl ether or 2-methyl-1-(methylbicyclo(2.2.1)-hept-5-en-2-yl)-1-penten-3-ol.

5. Claims 1-9 (partially), as far as the active ingredient is one or more of dibenzyl ether or methyl diphenyl ether, optionally with one or more of heliotropin or 2-methyl-1-(bicyclo(2.2.1)-hept-5-en-2-yl)-1-penten-3-ol.

6. Claims 1-9 (partially), as far as the active ingredient is heliotropin, optionally with 2-methyl-1-(methylbicyclo-(2.2.1 hept-5-en-2-yl)-1-penten-3-ol.

7. Claims 1-9 (partially), as far as the active ingredient is 2-methyl-1-(methylbicyclo(2.2.1)hept-5-en-2-yl)-1-penten-3-ol